Form PTO-1390US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE (Rev. 5-93)

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NO. **H3574 PCT/US**

U S APPLICATION NO (if known sec. 17 CFR 1 5) 09/856082

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/EP99/97532	**November 6, 1999**	**November 17, 1998**

TITLE OF INVENTION
LUBRICANTS FOR DRILLING FLUIDS

APPLICANT(S) FOR DO/EO/US
Heinz Mueller, Klaus-Peter Herold, Frank Bongardt, Nadja Herzog and Stephan von Tapavicza

Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:

1. ■ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.

2. ☐ This a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.

3. ☐ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).

4. ■ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.

5. ■ A copy of the International Application as filed (35 U.S. C. 371(c)(2)).
   a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ■ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).

6. ■ A translation of the International Application into English (35 U.S.C. 371(c)(2)).

7. ■ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ■ have not been made and will not be made.

8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).

9. ■ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). (UNEXECUTED)

10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.

12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.

13. ■ A FIRST preliminary amendment
    ☐ A SECOND or SUBSEQUENT preliminary amendment.

14. ☐ A substitute specification.

15. ☐ A change of power of attorney and/or address letter.

16. ■ Other items or information.:

Drawings (2 sheets)

"Express Mail" mailing label number EL541613196US

U.S. Application No. (if known see CFR1.30) 09/856082	INTERNATIONAL APPLICATION NO. PCT/EP99/08532	ATTORNEY'S DOCKET NUMBER H3574 PCT/US

17. ■ The following fees are submitted:

Basic National Fee (37 CFR 1.492(a)(1)-(5)):

Search Report has been prepared by the EPO or JPO..... . **$860.00**

International preliminary examination fee paid to USPTO (37CFR 1.482) **$690.00**

No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37CFR 1.445(a)(2))..... **$760.00**

Neither international preliminary examination fee (37CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO. **$1000.00**

International preliminary examination fee paid to USPTO (37CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)..... **$96.00**

	CALCULATIONS	PTO USE ONLY
ENTER APPROPRIATE BASIC FEE AMOUNT =	$ 860	00
Surcharge of $130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date 37 (CFR 1.492(e)).	$	

Claims	Number filed	Number Extra	Rate		
Total Claims	13 - 20 =	0	X 18.00	$ 0	00
Independent Claims	1 - 3 =	0	X 80.00	$ 0	00
Multiple dependent claims (s)(if applicable)		0	+ 260.00	$ 0	00
TOTAL OF ABOVE CALCULATIONS =				$ 860	00
Reduction by ½ for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).				$	
SUBTOTAL =				$ 860	00
Processing fee of **$130.00** for furnishing the English translation later the ☐ 20 ☐ 30 months from the earliest claimed priority date (37CFR 1.492(f))..... +				$	
TOTAL NATIONAL FEE =				$ 860	00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). $40.00 per property +				$	
TOTAL FEES ENCLOSED =				$ 860	00
				Amount to be: refunded	$----------
				charged	860.00

a. ☐ A check in the amount of $_____ to cover the above fees is enclosed.

b. ■ Please charge my Deposit Account No. 50-1177 in the amount of $ 860.00 to cover the above fees. A triplicate copy of this sheet is enclosed. Order No. 01-0302.

c. ■ The Assistant Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 50-1177. A triplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO: Cognis Corporation, Law Dept.
2500 Renaissance Blvd., Ste. 200
Gulph Mills, PA 19406

_____
SIGNATURE

Steven J. Trzaska
NAME ATTORNEY FOR APPLICANT

36,296
REGISTRATION NUMBER

Form PTO 1390 (REV 5-93)

"Express Mail" mailing label number EL541613196US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant	:	Mueller et al.
I.A. Number	:	PCT/EP99/08532
I.A. Filing Date:		November 6, 1999
Priority Date	:	November 17, 1998
Title	:	LUBRICANTS FOR DRILLING FLUIDS
Grp./A.U.	:	Unknown
Examiner	:	Unknown
Docket No.	:	H 3574 PCT/US

Assistant Commissioner for Patents
Box PCT
Washington, DC 20231

ATTN: DO/EO/US

PRELIMINARY AMENDMENT

Sir:

Preliminary to examination, please amend the instant application as follows.

In the Specification:

At page 1, before line 1, insert --Background of the Invention--.

At page 3, before line 19, insert

--Brief Description of the Several Views of the Drawing

Figure 1 is a graph comparing the coefficient of friction of the water-based fluid (I) without a lubricant and with the lubricant (b).

Figure 2 is a graph comparing the coefficient of friction of the oil-based fluid (II) without a lubricant and with the lubricant (b) + (c).--

At page 3, line 19, insert --Detailed Description of the Invention--.

Enter a new page 13, submitted herewith, containing the Abstract of the Disclosure.

In the Claims:

Cancel claims 1-10, without prejudice.

Please enter the following new claims.

11. A process for imparting lubricity to an aqueous drilling fluid used in geological exploration comprising:

(a) providing an aqueous drilling fluid;

(b) providing a lubricant component containing a partial glyceride of predominantly unsaturated fatty acids having from about 16 to 24 carbon atoms; and

(c) introducing the partial glyceride into the aqueous drilling fluid.

12. The process of claim 11 wherein the partial glyceride has a pour point of up to 10°C.

13. The process of claim 11 wherein the partial glyceride has a pour point of up to 0°C.

14. The process of claim 11 wherein the partial glyceride is derived from tall oil fatty acid.

15. The process of claim 11 wherein from about 0.5 to 5% by weight of the lubricant component, based on the weight of the drilling fluid, is introduced into the drilling fluid.

16. The process of claim 11 wherein the drilling fluid contains in excess of 90% by weight, based on the weight of the drilling fluid, of water.

17. The process of claim 11 wherein the drilling fluid contains in excess of 10% by weight, based on the weight of the drilling fluid, of a water-insoluble oil.

18. The process of claim 11 wherein the partial glyceride contains less than about 15% by weight, based on the weight of partial glyceride, of triglycerides.

19. The process of claim 11 wherein the partial glyceride contains less than about 5% by weight, based on the weight of partial glyceride, of triglycerides.

20. The process of claim 11 wherein from about 1 to 3% by weight of the lubricant component, based on the weight of the drilling fluid, is introduced into the drilling

fluid.

21. The process of claim 11 wherein the lubricant component further comprises an anionic surfactant.

22. The process of claim 21 wherein the anionic surfactant is selected from the group consisting of sulfonates, sulfates, and mixtures thereof.

23. The process of claim 21 wherein the anionic surfactant and the partial glyceride are employed in a ratio by weight of from about 1:10 to 1:20.

REMARKS/ARGUMENTS

Claims 11-23 are currently pending in the instant application.

The Specification has been amended to include the preferred section headings pursuant to 37 C.F.R. §1.77. An Abstract of the Disclosure has been added on a separate sheet. All of the amendments to the Specification constitute deletions of original section headings and/or paragraphs, and insertions or additions of new section headings and/or paragraphs. It is submitted that the amendments to the Specification made herein introduce no new matter. Their entry is therefore proper and respectfully requested. Accordingly, pursuant to 37 C.F.R. §1.121(b)(1)(iii), no separate page captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE" is necessary.

Original claims 1-10 have been canceled and replaced with new claims 11-23 solely for the purpose of improving clarity and grammar, which may suffer in translation, and not for any reason which relates to the statutory requirements for a patent. New claims 11-23 have not been added in response to any rejection, nor in anticipation of any rejection. Applicant(s) respectfully submit(s) that the scope of new claims 11-23 corresponds to the scope of original claims 1-10, and that new claims 11-23 are no narrower than original claims 1-10. Furthermore, although a moot point in view of their cancellation, Applicant(s) respectfully submit(s) that original claims 1-10 satisfied the requirements of 35 U.S.C. §112, as filed. New claims 11-23 are supported by the claims as originally filed and by the Examples. No new matter has been introduced. Entry is therefore proper and respectfully requested.

Prompt examination of the instant application in view of the

amendments made herein is respectfully requested.

Respectfully submitted,

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Attached:

1. Added Page 13

Abstract of the Disclosure

A process for imparting lubricity to an aqueous drilling fluid used in geological exploration involving: (a) providing an aqueous drilling fluid; (b) providing a lubricant component containing a partial glyceride of predominantly unsaturated fatty acids having from about 16 to 24 carbon atoms; and (c) introducing the partial glyceride into the aqueous drilling fluid.

unsaturated, linear or branched fatty acids containing 1 to 24 carbon atoms,

b) mono- and polyhydric, linear or branched alcohols containing 6 to 36 carbon atoms,

c) mineral oil, diesel oil, paraffin oil,

d) linear alpha-olefins and derivatives thereof and internal olefins,

e) carbonic acid esters.

10. The use claimed in any of claims 1 to 9, characterized in that the partial glycerides are used in combination with sulfonates of C_{12-24} fatty acids.

Lubricant for Drilling Fluids

The present invention relates to the use of partial glycerides as lubricants in drilling fluids for geological exploration.

It is known that drilling fluids for sinking wells in rock and bringing up the rock cuttings are flowable systems thickened to a limited extent which may be assigned to any of the following three classes: purely aqueous drilling fluids, oil-based drilling fluids, which are generally used as so-called invert emulsion fluids, and preparations of the w/o emulsion type in which the aqueous phase is heterogeneously distributed as a fine dispersion in the continuous oil phase. The third class of known drilling fluids is built up on water-based o/w emulsions, i.e. on liquid systems which contain a heterogeneous, finely disperse oil phase in a continuous aqueous phase.

In addition to the basic constituents of a drilling fluid, that is to say water and/or oil, such systems also contain a large number of further constituents which are essential to their performance properties. These include, for example, weighting agents, generally barium sulfate ("barite") which imparts the necessary density to the fluid. Water-soluble salts, generally calcium chloride, are also added in order to prevent an osmotic compensation between the formation water and the drilling fluid. To obtain stable emulsions, suitable emulsifiers are generally also added. Corrosion inhibitors, viscosity-regulating additives, fluid loss additives, alkali reserves and also lubricants may also be present in the fluids.

The class of pure water-based systems is the oldest in the historical development of drilling fluids. However, their use is attended by such serious disadvantages that, hitherto, only limited application has been possible for technically demanding drilling operations. Above all, the interaction of the water-based drilling fluids with the water-sensitive layers of rock - more particularly corresponding layers of clay - to be drilled leads

to unacceptable interference with the drilling process.

Very recently, however, a fairly old proposal has been taken up again and, even in highly sensitive shale formations, can lead to adequate stability where purely water-based drilling fluids are used. It involves the use of corresponding systems based on soluble alkali metal silicates which are also known as waterglasses or waterglass-based systems, cf. for example the seminar on "**The Prevention of Oil Discharge from Drilling Operations**" held in public in Aberdeen on 18th/19th June 1996 (organized by IBC Technical Services, London) and, in particular, the publications appearing in this connection by M. Eigner entitled "**Field Trials with a Silicate Drilling Fluid in Shell-Expro**" and by I. Ward and B. Williamson entitled "**Silicate Water Based Muds - a Significant Advance in Water Based Drilling Fluid Technology**".

However, the use of purely water-based drilling fluids, particularly of the last-mentioned type, makes the presence of components with a lubricating effect desirable. A large number of lubricants are known for use in practice. These include mineral oils, animal and vegetable oils and esters. The increasingly stricter regulations with regard to the biodegradability of drilling fluids and their constituents are gradually restricting the use of the otherwise particularly suitable mineral oils. At the same time, there is a growing interest in alternatives with better biodegradability, in particular esters. **EP 0 770 661**, for example, describes esters of monocarboxylic acids with monohydric alcohols as suitable lubricants for water-based drilling fluid systems. However, only a 2-ethylhexyl oleate is actually mentioned as a suitable lubricant for silicate-containing aqueous fluids. Applicants' **DE 196 47 598** describes C_{12-30} fatty alcohols and a mixture thereof with fatty acid esters as suitable lubricants for purely water-based silicate-containing drilling fluids. Triglycerides of fatty acids are also mentioned as esters. It is known that a particularly pronounced lubricating effect is attributed in particular to carboxylic acid

esters for the purpose of geological exploration, use being made of this in many ways. However, their use in water-based systems and above all in comparatively highly alkaline waterglass systems can lead to considerable difficulties. Ester cleavage can result in the formation of components with a marked tendency to foam which then introduce unwanted problems into the fluid systems. The above-mentioned publication by I. Ward et al. expressly refers to this difficulty.

Sulfonates of vegetable oils, in particular soya oil sulfonate, are also used as lubricants in practice. Soya oil sulfonate can be used in water- and oil-based systems, but shows significant foaming, especially in water-based fluids, which restricts its usefulness.

Accordingly, the problem addressed by the present invention was to provide a lubricant for both water- and oil-based drilling fluids which would not have the above-mentioned disadvantages. In addition, the lubricant would even be usable at low temperatures which can occur, for example, in drilling fluids that are used and stored in arctic regions. It has now been found that certain selected fatty acid partial glycerides have the desired properties.

In a first embodiment, therefore, the present invention relates to the use of partial glycerides of predominantly unsaturated C_{16-24} fatty acids, optionally in admixture with anionic surfactants, as lubricants in drilling fluids for geological exploration which contain water and optionally a separate oil phase.

It has proved to be advantageous to use partial glycerides which have a pour point - as measured to **DIN ISO 3016** - of at most 10°C and preferably at most 0°C.

The lubricants are selected from the partial esters of glycerol with predominantly unsaturated fatty acids containing 16 to 24 carbon atoms. Unsaturated fatty acids are understood to be carboxylic acids which contain at least one olefinically unsaturated double bond in the carbon chain.

However, polyunsaturated, in particular di- and tri-unsaturated, fatty acids are also suitable. These are mono- and/or diglycerides of fatty acids or, more particularly, fatty acid mixtures which contain unsaturated fractions to a predominant extent, i.e. more than 50% by weight. Such unsaturated fatty acids are, for example, palmitoleic acid, oleic acid, ricinoleic acid, linoleic acid, linolenic acid or arachidonic acid. Due to the process used for their production, the glycerides are generally not present as pure individual substances, but rather as mixtures of various partial glycerides with different fatty acids. These glycerides may be of natural origin or may be synthesized. Glyceride mixtures which contain at least 50% by weight of monoglycerides are particularly preferred. The percentage content of triglycerides in the partial glycerides used in accordance with the invention is less than 15% by weight, preferably less than 10% by weight and in particular less than 5% by weight.

Mixtures of mono- and diglycerides containing about 40 to 50% by weight of monoglycerides and the same percentage of diglycerides are generally used. The balance to 100% by weight consists of triglyceride.

Depending on the nature of the fatty acid mixture, the partial glycerides may also contain small amounts of saturated fatty acids from the group consisting of saturated C_{16-24} fatty acids. The use of partial glycerides of tall oil fatty acids, a mixture of 45 to 65% by weight of linoleic and conjugated C_{18} fatty acids, 25 to 45% by weight of oleic acid, 5 to 12% by weight of 5,9,12-octadecatrienoic acid and 1 to 3% by weight of saturated fatty acids (according to **Römpps Chemie Lexikon, 9th Edition, Volume 6, page 4,484, 1992**), is particularly preferred. Tall oil fatty acids are obtained by distillation from tall oil and are then esterified with glycerol on an industrial scale.

According to the invention, the partial glycerides may be used as lubricants both in water-based and in oil-based drilling fluid systems. Water-based systems generally contain only water as the base liquid,

preferably more than 90% by volume water. However, they may also contain water-insoluble oils, which form a separate oil phase, in quantities of 1 to 10% by volume. These fluids are then generally present in the form of an oil-in-water emulsion, suitable emulsifiers preferably being used in such cases. In the oil-based systems, the liquid phase of the fluid contains more than 10% by volume of water-insoluble oils. Preferred ratios by volume between oil and water are in the range from 90/10 to 60/40. Systems such as these form water-in-oil emulsions, optionally with the aid of suitable emulsifiers.

The partial glycerides are suitable as a lubricant component both for water-based and for oil-based fluids. In the case of water-based fluids in particular, the use of the partial glycerides leads to significantly reduced foaming which in turn facilitates adjustment of the viscosity and density in practice because, with foaming systems, the accurate measurement and adjustment of density is only possible with difficulty. However, the partial glycerides may also advantageously be used as lubricants in oil-based systems and, here also, lead to a significant reduction in the coefficient of friction, especially under the effect of pressure.

In the process according to the invention, the partial glycerides are added to the drilling fluids in quantities of 0.5 to 5% by weight, based on the drilling fluid as a whole, preferably in quantities of 0.5 to 3% by weight and more preferably in quantities of 1 to 3% by weight. Depending on the particular application and the fluid system, it may be necessary to add more lubricant during the actual drilling operation.

According to the invention, the partial glycerides are used as lubricants in drilling fluids which contain water and optionally a water-insoluble oil in the form of a separate oil phase. This water-insoluble oil is preferably chosen from the group consisting of

a) esters of monohydric saturated or unsaturated, linear or branched alcohols containing 1 to 24 carbon atoms and monobasic saturated or

unsaturated, linear or branched fatty acids containing 1 to 24 carbon atoms

b) mono- and polyhydric, linear or branched alcohols containing 6 to 36 carbon atoms

c) mineral oil, diesel oil, paraffin oil

d) linear alpha-olefins and derivatives thereof and internal olefins

e) carbonic acid esters.

Although mineral and diesel oils and the paraffin oils, particularly those containing 14 to 16 carbon atoms, are well-known oil phases for drilling fluids, ecologically safe alternatives have been increasingly developed in recent years. Because of their ready biodegradability, the esters in particular have become a tried and tested practical alternative to the pure hydrocarbons. Suitable ester oils are described in applicants' European patents **EP 0 374 671, EP 0 374 672, EP 0 386 638, EP 0 386 636** and **EP 0 535 074** of which the disclosures are also part of the present invention. Certain water-insoluble alcohols are also suitable oils for the drilling fluid systems in question here. Polyhydric, in particular dihydric, alcohols are preferably used. Water-soluble alcohols may also be mixed with water-insoluble solvents, particularly fatty acid esters, and the resulting mixture used as the oil phase. Applicants have described this class of compounds in detail in their European patents **EP 0 391 252** and **EP 0 472 558**. Another group of suitable oils are the linear alpha-olefins and derivatives thereof, in particular poly-alpha-olefins (PAOs). Suitable compounds of this type are described, for example, in the International patent application **WO 95/34610**. Internal olefins may also be used as oils for the purposes of the present invention. The carbonic acid esters described in Applicants' **EP 0 532 570** are also suitable base oils for the drilling fluid systems in question here. In principle, the above-mentioned oils by may be used individually or in admixture. Drilling fluid systems in which ecologically safe base oils, particularly esters or alcohols or mixtures

thereof, are used are particularly preferred.

Water-based drilling fluids and the additives to be used therein, such as weighting agents, fluid loss additives, alkali reserves, viscosity regulators and the like, are the subject matter of extensive general literature and relevant patent literature. Detailed technical information can be found, for example, in the book by George R. Gray and H.C.H. Darley entitled **"Composition in Properties of Oil Well Drilling Fluids", 4th Edition, 1980/81, Gulf Publishing Company Houston** and the extensive technical and patent literature cited therein and in the handbook entitled **"Applied Drilling Engineering"** by Adam T. Borgoyne, Jr. et al., **First Printing Society of Petroleum Engineers, Richardson, Texas (USA).**

The field of water-based silicate fluids also addressed by the teaching according to the invention is also general technical knowledge, cf. in particular the relevant above-mentioned publications. Suitable alkali metal silicates are, in particular, water-soluble sodium silicate and/or water-soluble potassium silicate with modulus values (molar ratio of SiO_2 to Na_2O or K_2O) of 1.0 to 3.3 and preferably 1.5 to 2.5. Preferred concentrations of the alkali metal silicates in the water-based fluids are at most about 10% by weight, preferably in a somewhat lower range, for example from about 3 to 8% by weight and more particularly in the range from about 4 to 7% by weight. Together with the alkali metal silicates, high concentrations of soluble nonreactive salts are normally intended to be present in the water-based liquid phases. Alkali metal chlorides, especially sodium chloride and/or potassium chloride, are particularly suitable. Correspondingly salt-saturated aqueous silicate fluids are preferred in practice. The pH values of these liquid phases are - comparatively - in the strongly alkaline range and are above pH 10 and more particularly are at least pH 11. It is this class of highly alkaline silicate fluids described here in which the lubricants according to the invention have proved to be particularly effective as an additive in small quantities of, for example, 1 to 3% by weight - based on

the fluid as a whole. For the industrial use of these purely water-based systems which has now become interesting again, it is possible to achieve a substantial increase in performance - as is known in principle for the use of components having a lubricating effect in water-based systems - by using very small quantities of organic components selected in accordance with the present invention. The lubricants according to the invention may also be used in water-based fluids containing glycols, particularly ethylene glycol, propylene glycol or butylene glycol and polymers thereof. Such systems - known to the expert as glycol fluids - contain up to 30% by weight of the above-mentioned glycols. The lubricants may also be used in water-based fluids containing silicates and glycols.

Oil-based drilling fluids are generally used as so-called invert emulsion muds which consist of a three-phase system, namely: oil, water and fine-particle solids. These are preparations of the w/o emulsion type, i.e. the aqueous phase is heterogeneously distributed as a fine dispersion in the continuous oil phase. Various additives are present for stabilizing the system as a whole and for establishing the desired performance properties including, in particular, emulsifiers or emulsifier systems, weighting agents, fluid loss additives, alkali reserves, viscosity regulators and the like. Relevant particulars can be found, for example, in the articles by P.A. Boyd et al. entitled **"New Base Oil Used in Low-toxicity Oil Muds", Journal of Petroleum Technology, 1985, 137 to 142** and by R.B. Bennett entitled **"New Drilling Fluid Technology - Mineral Oil Mud", Journal of Petroleum Technology, 1984, 975 to 981** and the literature cited therein. Particularly preferred emulsifier systems for use in invert drilling muds are described in **DE 196 43 840** of which the disclosure is also part of the present application.

Besides partial glycerides, it has proved to be of advantage to use surface-active compounds from the class of anionic surfactants. The sulfonates and sulfates are of particular importance in his regard. Suitable

compounds of this type are, for example, C_{9-13} alkyl benzene sulfonates, olefin sulfates, C_{12-18} alkane sulfates, alpha-sulfofatty acids, alkyl sulfates, alcohol sulfates and ether sulfates and also alkyl sulfosuccinic acids. According to the invention, sulfonates of fatty acids containing 12 to 24 carbon atoms and, more particularly, 12 to 14 carbon atoms are preferably used. Sulfated castor oil is also preferably used as an anionic surfactant. In cases where anionic surfactants are also used, they are employed in quantity ratios of anionic surfactants to partial glycerides of 1:10 to 1:20.

The use of selected partial glycerides in drilling fluids in accordance with the invention leads to reduced friction both in water-based and in oil-based systems. No harmful foaming occurs in water-based drilling fluids. The partial glycerides may also be used at low temperatures and are biodegradable. In addition, the partial glycerides used in accordance with the invention are not toxic in aquatic systems.

Examples

In the following Examples, the lubricating effect of water- and oil-based drilling fluids under various pressures was measured by the Almen-Wieland test.

The following drilling fluid systems (I) and (II) were tested:

(I) Water-based fluid		(II) Oil-based fluid (o/w ratio 75/25)	
Water	4 l	Mineral oil	675 ml
XC polymer	20 g	Water	225 ml
Bentonite	56 g	$CaCl_2$	95 g
CMC LVT	40 g	Emulsifier	35 g
Barite	1,755 g	Fluid loss additive	10 g
		Viscosifier	25 g
		Lime	17 g
		Barite	360 g

1.5% by weight of lubricant was added to each of the fluids. The following lubricants were investigated:

a) soya oil sulfonate

b) glycerol monotalloate (pour point: -15°C, saponification value: 155, acid value: 1, iodine value: 120, OH value: 255)

c) a combination of glycerol monotalloate with sulfated castor oil.

The graphs in Fig. 1 show the coefficient of friction of the water-based fluid (I) under various pressures. A fluid with no lubricant ("blank mud") was investigated for comparison. It can be seen that the use of partial glycerides b) in accordance with the invention leads to very good lubricating properties of the fluid, especially in the lower pressure range. At the same time, the fluids according to the invention generate hardly any foam.

The graphs in Fig. 2 show the test results for the oil-based fluid (II) against the lubricant-free fluid and a combination of b) with c). It can again be seen that the use of partial glycerides leads to a significant reduction in friction.

CLAIMS

1. The use of partial glycerides of predominantly unsaturated C_{16-24} fatty acids, optionally mixed with anionic surfactants, as lubricants in drilling fluids for geological exploration which contain water and optionally a separate oil phase.

2. The use claimed in claim 1, characterized in that partial glycerides with a pour-point - as measured to DIN ISO 3061 - of at most 10°C are selected.

3. The use claimed in claim 1 or 2, characterized in that partial glycerides with a pour point - as measured to DIN ISO 3061 - of at most 0°C are selected.

4. The use claimed in claims 1 to 3, characterized in that mono- or diglycerides of predominantly unsaturated C_{16-24} fatty acids or mixtures of these partial glycerides are selected as the lubricant.

5. The use claimed in any of claims 1 to 4, characterized in that mono- and/or diglycerides of tall oil fatty acid are selected as the lubricant.

6. The use claimed in any of claims 1 to 5, characterized in that drilling fluids containing the lubricant in quantities of 1 to 3% by weight, based on the drilling fluid as a whole, are used.

7. The use claimed in any of claims 1 to 6, characterized in that the lubricants are used in drilling fluids which contain more than 90% by volume of water.

8. The use claimed in any of claims 1 to 6, characterized in that the lubricants are used in drilling fluids which contain more than 10% by volume of a water-insoluble oil.

9. The use claimed in any of claims 1 to 8, characterized in that the lubricants are used in drilling fluids which contain water-insoluble oils selected from the group consisting of

a) esters of monohydric saturated or unsaturated, linear or branched alcohols containing 1 to 24 carbon atoms and monobasic saturated or

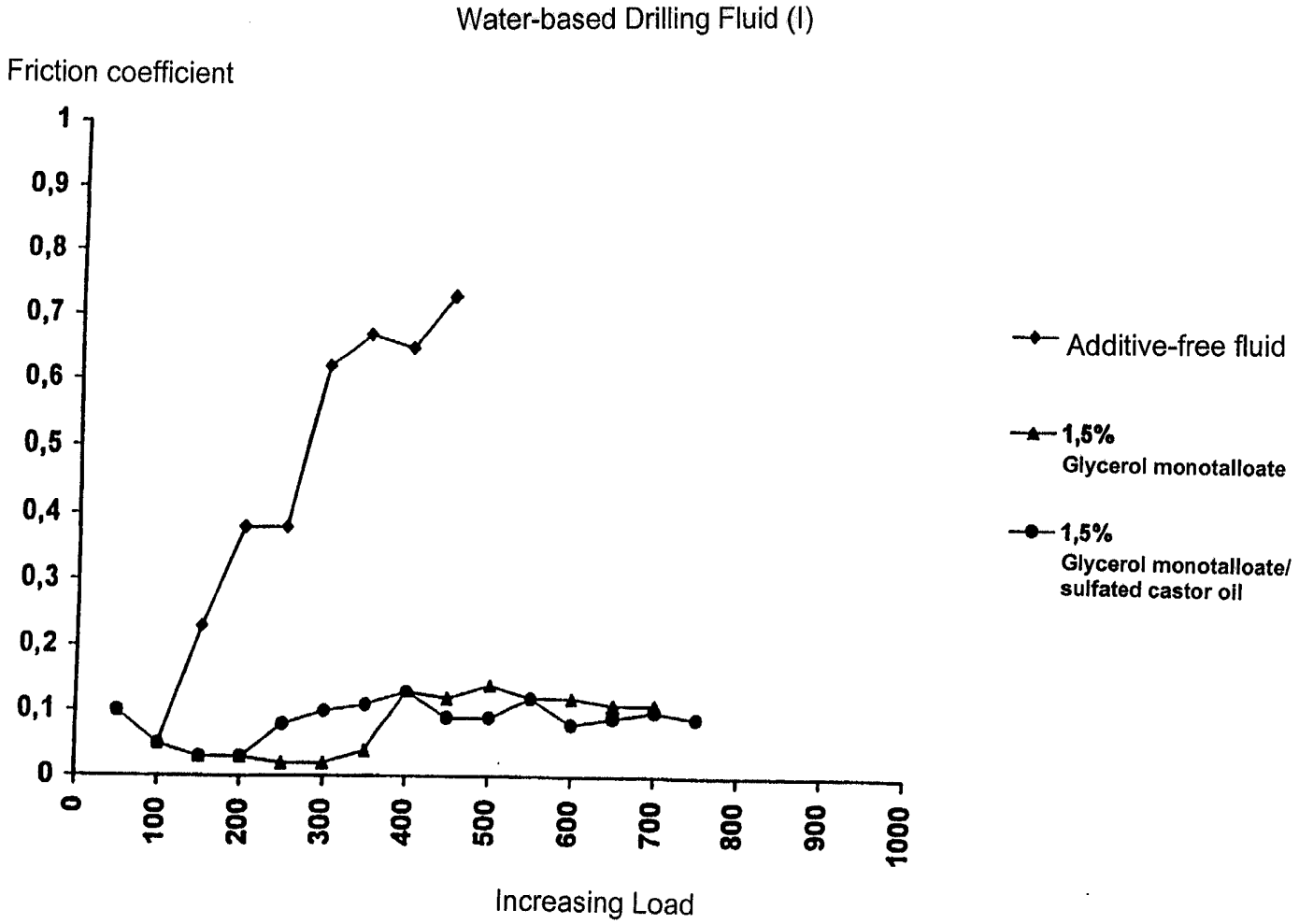

Fig. 1

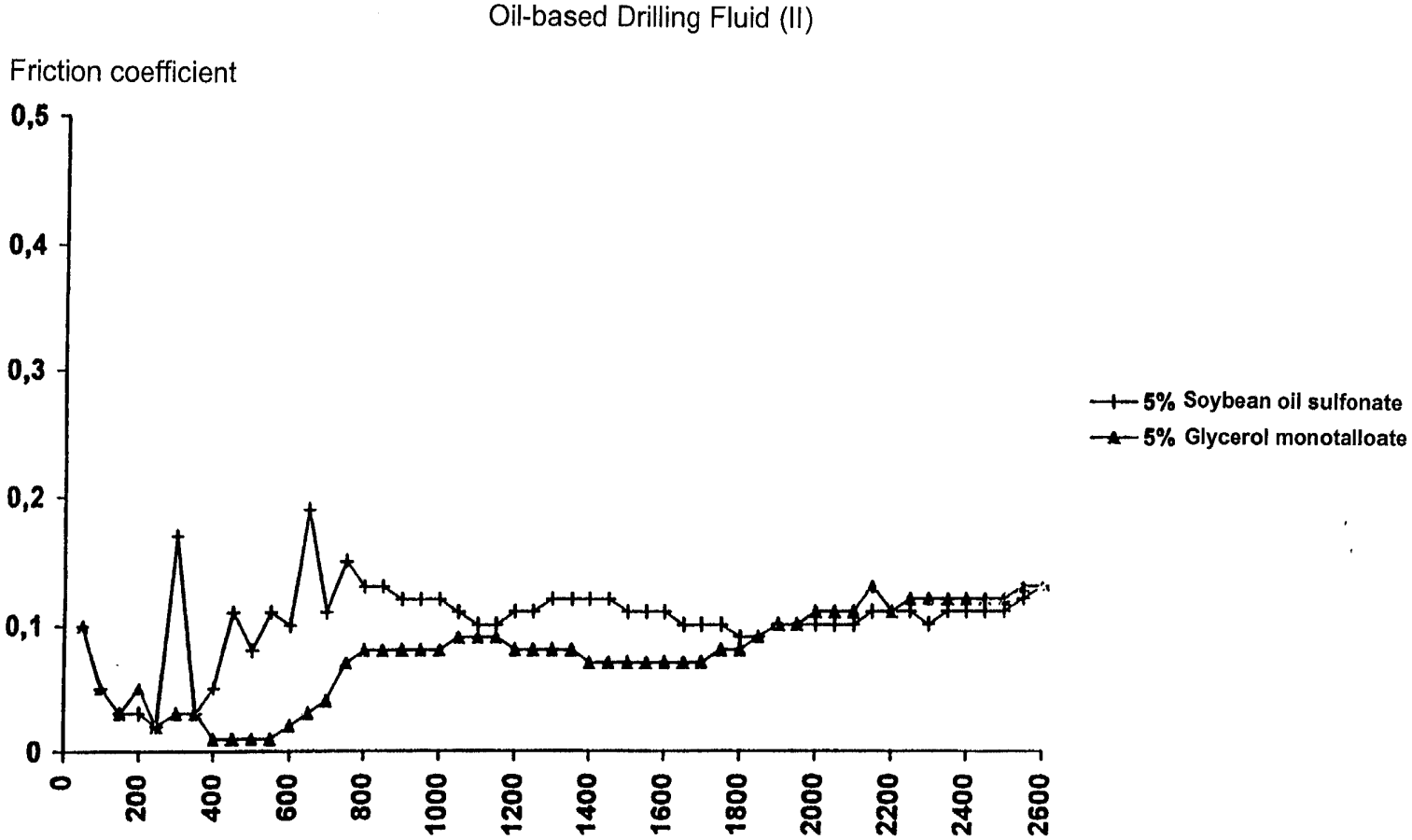

Fig. 2

"Express Mail" mailing label number EL541614157US

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PTO/SB/01 (6-95)
Approved for use through: 10/31/98 OMB 0651-0032
Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE

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DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION

☐ Declaration Submitted with Initial Filing OR ☒ Declaration Submitted after Initial Filing

Attorney Docket Number	H 3574 PCT/US
First Named Inventor	Mueller, Heinz
COMPLETE IF KNOWN	
Application Number	09/856,082
Filing Date	08/27/01
Group Art Unit	
Examiner Name	

As a below named inventor, I hereby declare that:
My residence, post office address, and citizenship are as stated below next to my name
I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

LUBRICANTS FOR DRILLING FLUIDS

(Title of the Invention)

the specification of which
☐ is attached hereto
OR
☒ was filed on (MM/DD/YYYY) 11/06/1999 as United States Application Number or PCT International Application Number PCT/EP99/08532 and was amended on (MM/DD/YYYY) _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1 56

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES	Certified Copy Attached? NO
198 52 971.6	Germany	11/17/1998	☐	☐	☒

☐ Additional foreign application numbers are listed on a supplemental priority sheet attached hereto.

I hereby claim the benefit under Title 35, United States Code §119(e) of any United States provisional application(s) listed below

Application Number(s)	Filing Date (MM/DD/YYYY)	Additional provisional application numbers are listed on a supplemental priority sheet attached hereto
		☐

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DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365© of any PCT international application designating the United Sates of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112 1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1 56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number *(if applicable)*
	PCT/EP99/08532	11/06/1999	

☐ Additional U S or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

☐ Firm Name OR ☐ Customer Number or label

☒ List Attorney(s) and/or agent(s) name and registration number below

Name	Registration Number	Name	Registration Number
John E. Drach	32,891	Steven J. Trzaska	36,296
Aaron E. Ettelman	42,516	Henry E. Millson, Jr.	18,980

☐ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

Please direct all correspondence to: ☒ Customer Number or label **23657** OR ☐ Fill in correspondence address below

Name	Steven J. Trzaska				
Address					
Address					
City		State		Zip	
Country		Telephone	610-278-4929	Fax	610-278-6548

hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this unsigned inventor

Given Name	**Heinz**	Middle Initial		Family Name	**Mueller**	Suffix e.g. Jr.	
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Post Office Address							
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Applicant Authority							

☒ Additional inventors are being named on supplemental sheet(s) attached hereto

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H 3574 PCT/US

DECLARATION

ADDITIONAL INVENTOR(S) Supplemental Sheet

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Claus-Peter	Middle Initial		Family Name	Herold	Suffix e.g. Jr.	
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Post Office Address							
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Applicant Authority							

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

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Post Office Address							
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Applicant Authority							

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Nadja	Middle Initial		Family Name	Herzog	Suffix e.g. Jr.	
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Post Office Address							
City	40699 Erkrath	State		Zip		Country	Germany
Applicant Authority							

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Stephan	Middle Initial		Family Name	von Tapavicza	Suffix e.g. Jr.	
Inventor's Signature					Date	07/05/2001	
Residence: City	Erkrath	State		Country	Germany DEX	Citizenship	Germany
Post Office Address	Thomas-Mann-Strasse 12						
Post Office Address							
City	40699 Erkrath	State		Zip		Country	Germany
Applicant Authority							

☐ Additional inventors are being named on supplemental sheet(s) attached hereto